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IN THE UNITED STATES PATENT & TRADEMARK OFFICE

IN RE APPLICATION OF

HIROYUKI KISO, ET AL. : EXAMINER: COONEY, J. M.

SERIAL NO: 10/618,693

FILED: JULY 15, 2003 : GROUP ART UNIT: 1711

RCE FILED: NOVEMBER 23, 2005

FOR: CATALYST AND PROCESS FOR PRODUCING A RIGID POLYURETHANE

FOAM

CORRECTED APPEAL BRIEF

COMMISSIONER FOR PATENTS ALEXANDRIA, VIRGINIA 22313

SIR:

This is a corrected Appeal Brief, in response to the Notification of Non-Compliant Appeal Brief mailed February 2, 2007.

Applicants submit that while only the corrected non-compliant sections are required to be filed, it is more beneficial to simply file a corrected Appeal Brief, which corrects the non-compliant sections.

This is an appeal of the Final Rejection dated July 11, 2006 of Claims 1, 4, 29-33, 37 and 38. A Notice of Appeal was timely filed on October 10, 2006.

I. REAL PARTY IN INTEREST

The real party in interest in this appeal is Tosoh Corporation having an address 4560, Kaisei-cho, Shunan-shi, Yamaguchi-ken, 746-8501, Japan.

II. RELATED APPEALS AND INTERFERENCES

Appellants, Appellants' legal representative and the assignee are aware of no appeals, interferences, or judicial proceedings which may be related to, directly affect or be directly affected by or have a bearing on the Board's decision in this appeal.

III. STATUS OF THE CLAIMS

Claims 1, 4, 29-33, 37 and 38 stand rejected and are herein appealed. Claims 5-28 stand withdrawn from consideration. Claims 2, 3, and 34-36 have been canceled.

IV. STATUS OF THE AMENDMENTS

An amendment under 37 CFR 41.33 was filed on December 6, 2006. An Advisory Action entered February 2, 2007 indicated that the amendment would not be entered. A corrected amendment under 37 CFR 41.33 is submitted herewith, which cancels Claims 34-36 and rewrites Claim 37 in independent form, without change in claim scope. The Claims Appendix reflects the claims as if the corrected amendment were entered.

V. SUMMARY OF THE CLAIMED SUBJECT MATTER

Independent Claim 1 is drawn to a catalyst for producing a rigid polyurethane foam by means of at least one blowing agent selected from the group consisting of 1,1,1,3,3pentafluoropropane (HFC-245fa), 1,1,1,3,3-pentafluorobutane (HFC-365mfc) and a low boiling point hydrocarbon, which comprises:

at least one amine compound (1) selected from the group consisting of dimethylethylamine, dimethylpropylamine, dimethylbutylamine, dimethylpentylamine, dime

and at least one amine compound (2) selected from the group consisting of triethylenediamine, N,N,N',N'-tetramethyl-1,6-hexanediamine and N,N-dimethylcyclohexylamine,

wherein said at least one amine compound (1) and said at least one amine compound (2) are present in amounts effective to improve at least one property of a rigid polyurethane foam produced by means of said at least one blowing agent and said catalyst, compared to an otherwise identical rigid polyurethane foam produced using said at least one amine compound (1) alone, or said at least one amine compound (2) alone, as the catalyst, and wherein said at least one property includes dimensional stability.

Independent Claim 37 is drawn to a catalyst for producing a rigid polyurethane foam by means of at least one blowing agent selected from the group consisting of 1,1,1,3,3-pentafluoropropane (HFC-245fa), 1,1,1,3,3-pentafluorobutane (HFC-365mfc) and a low boiling point hydrocarbon, which comprises:

at least one amine compound (1) selected from the group consisting of dimethylbutylamine, dimethylbutylamine, dimethyloctylamine, and dimethyldodecylamine,

and at least one amine compound (2) selected from the group consisting of triethylenediamine, N,N,N',N'-tetramethyl-1,6-hexanediamine and N,N-dimethylcyclohexylamine,

wherein said at least one amine compound (1) and said at least one amine compound (2) are present in amounts effective to improve at least one property of a rigid polyurethane foam produced by means of said at least one blowing agent and said catalyst, compared to an otherwise identical rigid polyurethane foam produced using said at least one amine compound (1) alone, or said at least one amine compound (2) alone, as the catalyst, and wherein said at least one property includes dimensional stability.

Claim 37 is analogous to Claim 1, but it narrows the scope of the compound (1) Markush group.

Independent Claim 38 is drawn to a catalyst for producing a rigid polyurethane foam by means of at least one blowing agent selected from the group consisting of 1,1,1,3,3-pentafluoropropane (HFC-245fa), 1,1,1,3,3-pentafluorobutane (HFC-365mfc) and a low boiling point hydrocarbon, which comprises:

at least one amine compound (1) selected from the group consisting of dimethylethylamine, dimethylpropylamine, dimethylbutylamine, dimethylpentylamine, dime

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and at least one amine compound (2) selected from the group consisting of triethylenediamine, N,N,N',N'-tetramethyl-1,6-hexanediamine and N,N-dimethylcyclohexylamine,

wherein said at least one amine compound (1) and said at least one amine compound (2) are present in amounts effective to improve a balance of properties of a rigid polyurethane foam produced by means of said at least one blowing agent and said catalyst, compared to an otherwise identical rigid polyurethane foam produced using said at least one amine compound (1) alone, or said at least one amine compound (2) alone, as the catalyst, and wherein said balance of properties is a combination of flowability, adhesive strength and dimensional stability.

Claim 38 is analogous to Claim 1, but the functional amounts are in reference to a balance of the recited properties.

Support for the above independent claims appear in original Claims 1 and 3, together with the specification at page 13, lines 2-13 and page 19, lines 10-22, with support for the recited amounts in Claims 1, 37 and 38 at page 12, lines 5-12, together with the comparative data in the specification beginning at page 53, line 17.

VI. GROUNDS OF REJECTION

Ground (A)

Claims 1, 4, 29-33 and 38 stand rejected under 35 U.S.C. § 103(a) as unpatentable over U.S. 4,585,804 (Lancaster et al) alone, or in view of U.S. 4,742,089 (Naka et al).

Ground (B)

Claims 1, 4, 29-38 stand rejected under 35 U.S.C. § 103(a) as unpatentable over U.S. 4,742,089 (Canaday et al) alone, or in view of Naka et al.

Ground (C)

Claims 1, 29-33 and 36-38 stand rejected under 35 U.S.C. § 112, first paragraph, as failing to satisfy the description requirement thereof.

VII. ARGUMENT

Preface to Grounds (A) and (B)

The present invention is based on Applicants' discovery that the combination of amine compounds (1) and (2) of the present claims produces a polyurethane foam that is superior to one using either amine compound alone as a catalyst when used in conjunction with a blowing agent selected from the group consisting of HFC-245fa, HFC-365mfc, and a low boiling point hydrocarbon.

Two sets of comparative data in the specification evidence this superiority, as discussed and set forth below.

In the first set of comparative data, which demonstrates the significance of using HFC-245fa or HFC-365mfc as the blowing agent, Examples 1-24 are according to the presently-claimed invention, and Comparative Examples 1-29 are not. The only difference between Comparative Examples 1-14 and the Examples is the catalyst used. Comparative Examples 15-26 employ the amine combination of the present invention, but with HCFC-

141b as the blowing agent, i.e., a blowing agent other than HFC-245fa, HFC-365mfc, and a low boiling point hydrocarbon.

The data are shown in Tables 2, 4 and 6, at pages 58-59, 62, and 67, respectively. As described at page 68, lines 14-16, in each Example, a rigid urethane foam excellent in flowability, adhesive strength and dimensional stability, was obtained. For Comparative Examples 1-4 and 8-11, which used only amine (1) as the catalyst, large amounts of the catalyst were required, and the foams were inferior in flowability and dimensional stability, as described at page 68, lines 17-23. For Comparative Examples 5-7 and 12-14, which used only amine (2) as the catalyst, the foams were inferior in flowability, adhesive strength and dimensional stability, as described at the paragraph bridging pages 68 and 69. For Comparative Examples 15-26, using the amine combination of the present claims but with a different blowing agent caused little change in flowability, adhesive strength or dimensional stability of the foams.

The second set of comparative data is analogous to the above-discussed first set, but highlights the significance of using a low boiling point hydrocarbon, such as cyclopentane alone or combined with isopentane, as an applicable blowing agent. See Tables 8, 10 and 12, and the descriptions of Examples 25-48, and Comparative Examples 30-58, in the specification at page 81, lines 1-26.

The comparative data is set forth in greater detail below. In the Tables, as can be confirmed by, for example, Table 1 at page 57 of the specification, Catalyst A is dimethylbutylamine; Catalyst B is dimethylhexylamine; Catalyst C is dimethyloctylamine; Catalyst D is dimethyldodecylamine; Catalyst E is triethylenediamine; Catalyst F is N,N,N',N'-tetramethyl-1,6-hexanediamine; and Catalyst G is N,N-dimethylcyclohexylamine.

When HFC-245fa was used as a blowing agent:

Catalyst	Data	Flowability (cm)	Adhesive strength (kgf/cm²)	Dimensional stability (%)
Combination of				
Catalyst A and	Examples 1 to 3	81 to 82	1.17 to 1.20	-1.3 to -1.0
at least one of				
Catalysts E to G	,	•		
(Present Inv.)		·		
Catalyst A alone	Comparative	•		
	Example 1	76	1.38	-8.8
Catalyst E alone	Comparative			
	Example 5	72	0.45	-3.5
Catalyst F alone	Comparative			
	Example 6	76	0.51	-5.2
Catalyst G alone	Comparative			
	Example 7	76	0.48	-5.6

Catalyst	Data	Flowability (cm)	Adhesive strength (kgf/cm ²)	Dimensional stability (%)
Combination of		•		
Catalyst B and	Examples 4 to 6	79 to 81	1.32 to 1.45	-1.2 to -0.7
at least one of				
Catalysts E to G				
(Present Inv.)				·
Catalyst B alone	Comparative			
	Example 2	77	1.56	-9.2
Catalyst E alone	Comparative			
	Example 5	72	0.45	-3.5
Catalyst F alone	Comparative	٠.		
	Example 6	76	0.51	-5.2
Catalyst G alone	Comparative			
	Example 7	76	0.48	-5.6

Catalyst	Data	Flowability (cm)	Adhesive strength (kgf/cm²)	Dimensional stability (%)
Combination of	Examples 7 to 9	79 to 83	1.21 to 1.33	-1.2 to -0.8
Catalyst C and at least one of	Examples / to 9	79 10 83	1.21 to 1.33	-1.2 to -0.8
Catalysts E to G				
(Present Inv.)				
Catalyst C alone	Comparative			
	Example 3	75	1.5	-8.5
Catalyst E alone	Comparative			
	Example 5	72	0.45	-3.5
Catalyst F alone	Comparative	·		
	Example 6	76	0.51	-5.2
Catalyst G alone	Comparative			
	Example 7	76	0.48	-5.6

Catalyst	Data	Flowability (cm)	Adhesive strength (kgf/cm ²)	Dimensional stability (%)
Combination of Catalyst D and at least one of Catalysts E to G (Present Inv.)	Examples 10 to 12	79 to 83	1.14 to 1.28	-1.5 to -0.9
Catalyst D alone	Comparative Example 4	76	1.41	-7.7
Catalyst E alone	Comparative Example 5	72	0.45	-3.5
Catalyst F alone	Comparative Example 6	76	0.51	-5.2
Catalyst G alone	Comparative Example 7	76	0.48	-5.6

As is evident from the above tables, where HFC-245fa was used as a blowing agent, when Catalyst A, B, C or D was used alone, the dimensional stability was reduced substantially. Further, when Catalyst E, F or G was used alone, the adhesive strength and dimensional stability were reduced substantially. Accordingly, the adhesive strength and

dimensional stability can be improved by the combination of Catalyst A, B, C or D and Catalyst E, F or G in the present invention.

When HFC-365mfc was used as a blowing agent:

Catalyst	Data	Flowability (cm)	Adhesive strength (kgf/cm²)	Dimensional stability (%)
Combination of Catalyst A and at least one of Catalysts E to G (Present Inv.)	Examples 13 to 15	78 to 80	1.26 to 1.48	-1.2 to -0.9
Catalyst A alone	Comparative Example 8	75	1.62	-8.2
Catalyst E alone	Comparative Example 12	72	0.67	-2.7
Catalyst F alone	Comparative Example 13	75	0.76	-4.3
Catalyst G alone	Comparative Example 14	76	0.72	-4.4

Catalyst	Data	Flowability (cm)	Adhesive strength (kgf/cm ²)	Dimensional stability (%)
Combination of Catalyst B and at least one of Catalysts E to G	Examples 16 to 18	78 to 79	1.42 to 1.66	-1.1 to -0.7
(Present Inv.)				
Catalyst B alone	Comparative		÷ 2	
	Example 9	75	1.75	-7.5
Catalyst E alone	Comparative Example 12	72	0.67	-2.7
Catalyst F alone	Comparative Example 13	75	0.76	-4.3
Catalyst G alone	Comparative Example 14	76	0.72	-4.4

Catalyst	Data	Flowability (cm)	Adhesive strength (kgf/cm²)	Dimensional stability (%)
Combination of				
Catalyst C and	Examples	78 to 80	1.30 to 1.52	-1.2 to -0.7
at least one of	19 to 21			,
Catalysts E to G	•			
(Present Inv.)				
Catalyst C alone	Comparative	,		
	Example 10	75	1.68	-7.8
Catalyst E alone	Comparative			
	Example 12	. 72	0.67	-2.7
Catalyst F alone	Comparative	,		
	Example 13	75	0.76	-4.3
Catalyst G alone	Comparative	• .		
	Example 14	76	0.72	-4.4

Catalyst	Data	Flowability (cm)	Adhesive strength (kgf/cm²)	Dimensional stability (%)
Combination of Catalyst D and at least one of Catalysts E to G (Present Inv.)	Examples 22 to 24	78 to 81	1.25 to 1.43	-1.4 to -0.8
Catalyst D alone	Comparative Example 11	. 76	1.55	-7.2
Catalyst E alone	Comparative Example 12	72	0.67	-2.7
Catalyst F alone	Comparative Example 13	75	0.76	-4.3
Catalyst G alone	Comparative Example 14	76	0.72	-4.4

As is evident from the above tables, where HFC-365mfc was used as a blowing agent, when Catalyst A, B, C or D was used alone, the dimensional stability was reduced substantially. Further, when Catalyst E, F or G was used alone, the adhesive strength and dimensional stability were reduced substantially. Accordingly, the adhesive strength and

dimensional stability are improved by the combination of Catalyst A, B, C or D and Catalyst E, F or G in the present invention.

When a low boiling point hydrocarbon was used as a blowing agent:

Catalyst	Data	Flowability (cm)	Adhesive strength (kgf/cm ²)	Dimensional stability (%)
Combination of Catalyst A and at least one of	Examples 25 to 27, and 37 to 39	78 to 81	1.59 to 1.81	-1.2 to -0.8
Catalysts E to G (Present Inv.)				
Catalyst A alone	Comparative Examples 30 and 37	73 to 74	1.74 to 1.79	-7.7 to -6.8
Catalyst E alone	Comparative Examples 34 and 41	70	0.88 to 0.90	-3.1 to -2.8
Catalyst F alone	Comparative Examples 35 and 42	73 to 74	1.00 to 1.02	-4.6 to -4.3
Catalyst G alone	Comparative Examples 36 and 43	73 to 74	0.95 to 0.96	-5.0 to -4.3

Catalyst	Data	Flowability (cm)	Adhesive strength (kgf/cm ²)	Dimensional stability (%)
Combination of Catalyst B and at least one of	Examples 28 to 30, and 40 to 42	76 to 79	1.80 to 2.03	-1.1 to -0.5
Catalysts E to G (Present Inv.)	•			
Catalyst B alone	Comparative Examples 31 and 38	74 to 75	1.95 to 2.03	-8.1 to -7.2
Catalyst E alone	Comparative Examples 34 and 41	70	0.88 to 0.90	-3.1 to -2.8
Catalyst F alone	Comparative Examples 35 and 42	73 to 74	1.00 to 1.02	-4.6 to -4.3
Catalyst G alone	Comparative Examples 36 and 43	73 to 74	0.95 to 0.96	-5.0 to -4.3

Catalyst	Data	Flowability (cm)	Adhesive strength (kgf/cm²)	Dimensional stability (%)
Combination of Catalyst C and at least one of Catalysts E to G (Present Inv.)	Examples 31 to 33, and 43 to 45	76 to 81	1.64 to 1.86	-1.1 to -0.6
Catalyst C alone	Comparative Examples 32 and 39	.72 to 73	1.88 to 1.95	-7.5 to -6.8
Catalyst E alone	Comparative Examples 34 and 41	70	0.88 to 0.90	-3.1 to -2.8
Catalyst F alone	Comparative Examples 35 and 42	73 to 74	1.00 to 1.02	-4.6 to -4.3
Catalyst G alone	Comparative Examples 36 and 43	73 to 74	0.95 to 0.96	-5.0 to -4.3

Catalyst	Data	Flowability (cm)	Adhesive strength (kgf/cm ²)	Dimensional stability (%)
Combination of Catalyst D and at least one of Catalysts E to G (Present Inv.)	Examples 34 to 36, and 46 to 48	76 to 81	1.55 to 1.79	-1.4 to -0.7
Catalyst D alone	Comparative Examples 33 and 40	73 to 74	1.78 to 1.83	-6.8 to -6.0
Catalyst E alone	Comparative Examples 34 and 41	70	0.88 to 0.90	-3.1 to -2.8
Catalyst F alone	Comparative Examples 35 and 42	73 to 74	1.00 to 1.02	-4.6 to -4.3
Catalyst G alone	Comparative Examples 36 and 43	73 to 74	0.95 to 0.96	-5.0 to -4.3

As is evident from the above tables, where a low boiling point hydrocarbon was used as a blowing agent, when Catalyst A, B, C or D was used alone, the dimensional stability was reduced substantially. Further, when Catalyst E, F or G was used alone, the adhesive strength and dimensional stability were reduced substantially. Accordingly, the adhesive strength and dimensional stability can be improved by the combination of Catalyst A, B, C or D and Catalyst E, F or G in the present invention.

As described in the specification beginning at page 9, line 25, blowing agents of the type recited in the present claims are hardly soluble in a polyol as compared to dichloromonofluororethane (HCFC) analogs, are expensive, and if various tertiary amine catalysts are used, the amount of water to be used will be substantial, whereby there will be a problem that, compared to rigid polyurethane foams obtained using conventional HCFC or

the like as a blowing agent, the foam will be inferior in flowability, adhesive strength and dimensional stability. Thus, the presently-claimed invention is particularly efficacious when used to make rigid polyurethane foams using at least one of the blowing agents recited in the present claims.

Ground (A)

Claims 1, 4, 29-33 and 38 stand rejected under 35 U.S.C. § 103(a) as unpatentable over Lancaster et al alone, or in view of Naka et al. That rejection is untenable and should not be sustained.

Lancaster et al discloses "improved" rigid polyurethane foams produced in the presence of, *inter alia*, a blowing agent and at least one catalyst, wherein the improvement comprises incorporating a compound having at least one primary aliphatic amine group of a particular average molecular weight range (column 1, lines 35-56). Applicable catalysts listed include, *inter alia*, triethylene diamine and dimethylethylamine (paragraph bridging columns 3 and 4). While Lancaster et al discloses that mixtures of catalysts can be used, no advantage is disclosed from doing so.

Naka et al is drawn to the production of rigid polyurethane foams by reacting a polyol component having a special composition with an isocyanate component in the presence of a blowing agent, a reaction catalyst and a foam stabilizer having particular properties (Abstract). The Examiner relies on Naka et al's listing of triethylene diamine, as well as dimethylcyclohexylamine and tetramethylhexamethylene diamine, finding them to be "equivalent." Note, however, that among the blowing agents listed in Naka et al (column 4, lines 24-34), none are within the terms of the present claims.

The Examiner holds that it would have been obvious both to use the triethylene diamine and dimethylethylamine of Lancaster et al in combination, and to substitute the triethylene diamine with the "equivalent" dimethylcyclohexylamine or tetramethylhexamethylene diamine of Naka et al in that combination.

In reply, Naka et al does not disclose triethylene diamine, dimethylcyclohexylamine and tetramethylhexamethylene diamine, as necessarily "equivalent." Rather, they are each listed simply as applicable in Naka et al's reaction catalyst. More significantly, the applied prior art recognizes no benefit in using various catalysts in combination, compared to the use of individual catalysts alone.

Thus, the active claims are necessarily patentable, since they are inclusive only of catalysts which provide a rigid polyurethane foam having at least one improved property, as recited, such as dimensional stability, compared to the use of component (1) alone, or component (2) alone. It should not be necessary for Applicants to actually **show** superiority for all such compositions, since the prior art, as discussed above, recognizes no benefit from any combination of the above amines.

In sum, neither the presently-claimed invention, nor the above-discussed superior results, are disclosed or suggested by the applied prior art.

In the Final Rejection, the Examiner cites various precedent with regard to showings of unexpected results, and finds that Applicants have not demonstrated unexpected results in that they have not compared to the prior art relied on and that the showings are not commensurate in scope with the breadth of the claims.

In reply, what can be a more germane comparison when an invention is drawn to a combination of two components, where each component is known individually for a

particular utility, the prior art suggests no advantage in using them in combination, and the comparison is of the combination compared to each component alone? What prior art of record is closer? Indeed, the comparative data is more probative than the applied prior art since it relates to particular blowing agents recited in the claims. Compare *Ex parte Humber*, 217 USPQ 265 (Bd. Pat. App. & Inter. 1981) (comparative data showing the claimed chlorine-containing compounds to be unexpected over various (non-prior art) chlorine-containing isomers was accepted as more probative over prior art, drawn to non-chlorine containing analogs of the claimed compounds, asserted to be closest.) The applied prior art does not disclose these blowing agents.

With regard to whether the showing is commensurate in scope with the claims, the claims require that the relative amounts of the respective components (1) and (2) be in amounts effective to improve at least one property of a rigid polyurethane foam produced by means of a particular blowing agent and the particular combination of components (1) and (2), compared to identical subject matter but using component (1) alone or component (2) alone, and wherein the at least one property includes dimensional stability. The Examiner would presumably require submission of data for compositions of all possible relative amounts of components (1) and (2), suggesting that there are some combinations of component (1) and component (2) that do not result in an improved property, such as amounts wherein one of the components is present in an infinitesimal amount.

If that is the Examiner's position, Applicants submit that since they have already shown that there are such compositions that do result in an improvement in property, it would be a matter of routine experimentation to determine numerical percentage ranges in which such improved properties are obtained. It would defeat the Constitutional goal of advancing

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progress in the useful arts to delay early disclosure of a meritorious invention by conducting nothing more than routine experimentation.

Claim 29

Claim 29 is separately patentable, because the combination of <u>Lancaster et al</u> and <u>Naka et al</u> could not have predicted the above-discussed superior results when using triethylenediamine as amine compound (2).

Claim 30

Claim 30 is separately patentable, because the combination of <u>Lancaster et al</u> and <u>Naka et al</u> could not have predicted the above-discussed superior results when using N,N,N',N'-tetramethyl-1,6-hexanediamine as amine compound (2).

Claim 31

Claim 31 is separately patentable, because the combination of <u>Lancaster et al</u> and <u>Naka et al</u> could not have predicted the above-discussed superior results when using N,N-dimethylcyclohexylamine as amine compound (2).

Claim 38

Claim 38 is separately patentable, since the applied prior art does not suggest catalysts which provide a rigid polyurethane foam having an improved balance of flowability, adhesive strength and dimensional stability, compared to the use of component (1) alone, or component (2) alone.

For all the above reasons, it is respectfully requested that the rejection be REVERSED.

Ground (B)

Claims 1, 4, and 29-38 stand rejected under 35 U.S.C. § 103(a) as unpatentable over Canaday et al alone, or in view of Naka et al. That rejection is untenable and should not be sustained.

Canaday et al discloses polyurethane foams derived from, *inter alia*, at least one halogenated organic blowing agent and at least one amine catalyst (column 2, lines 13-25). While Canaday et al discloses that any of the blowing agents generally employed in similar prior art foam products may be employed, Canaday et al lists only chlorine-containing blowing agents (paragraph bridging columns 10 and 11). As applicable amines, Canaday et al lists tertiary amines such as triethylenediamine and dimethylbutylamine (paragraph bridging columns 11 and 12). While Canaday et al discloses that mixtures of the amines may be used (column 12, lines 3-4), no advantage is disclosed from doing so.

Naka et al has been discussed above under Ground (A).

The Examiner, in effect, employs <u>Canaday et al</u> in the same way as <u>Lancaster et al</u> is applied, as discussed above under Ground (A). Applicants' response in Ground (A) is the same herein, except that <u>Canaday et al</u> replaces <u>Lancaster et al</u>.

Claim 29

Claim 29 is separately patentable, because the combination of <u>Canaday et al</u> and <u>Naka et al</u> could not have predicted the above-discussed superior results when using triethylenediamine as amine compound (2).

Claim 30

Claim 30 is separately patentable, because the combination of <u>Canaday et al</u> and <u>Naka et al</u> could not have predicted the above-discussed superior results when using N,N,N',N'-tetramethyl-1,6-hexanediamine as amine compound (2).

Claim 31

Claim 31 is separately patentable, because the combination of <u>Canaday et al</u> and <u>Naka et al</u> could not have predicted the above-discussed superior results when using N,N-dimethylcyclohexylamine as amine compound (2).

Claim 37

Claim 37 is separately patentable, because the combination of <u>Canaday et al</u> and <u>Naka et al</u> could not have predicted the above-discussed superior results when using any of the amines in the component (1) Markush group, together with any of the amines in the component (2) Markush group.

Claim 38

Claim 38 is separately patentable, since the applied prior art does not suggest catalysts which provide a rigid polyurethane foam having an improved balance of flowability, adhesive strength and dimensional stability, compared to the use of component (1) alone, or component (2) alone.

For all the above reasons, it is respectfully requested that the rejection be REVERSED.

Ground (C)

Claims 1, 29-33 and 36-38 stand rejected under 35 U.S.C. § 112, first paragraph, as failing to satisfy the description requirement therein. That rejection is untenable and should not be sustained.

The Examiner finds that the recital of the at least one amine compound (1) and the at least one amine compound (2) in functional effective amounts, as recited at the end of Claims 1 and 38, is not described in the specification.

In reply, the issue is whether Applicants had possession of the invention now claimed as of the filing date. Indeed, the epitome of the embodiment of the active claims is Applicants' discovery that when using any of the blowing agents recited in the claims, the recited catalyst, i.e., the combination of components (1) and (2), produces superior results not obtained by using component (1) alone, or component (2) alone. This discovery is borne out by the above-discussed comparative data of record, wherein Applicants actually describe the differences in results. See, for example, the description at page 68, lines 17-23, wherein

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deficiencies resulting from the use of amine compound (1) are described, and the following paragraph, i.e., bridging pages 68 and 69, wherein deficiencies resulting from the use of amine compound (2) alone are described, compared to the combination of components (1) and (2).

For all the above reasons, it is respectfully requested that the rejection be REVERSED.

VIII. CONCLUSION

For the above reasons, it is respectfully requested that all the rejections still pending be REVERSED.

Respectfully submitted,

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HAP/rle

CLAIMS APPENDIX

Claim 1: A catalyst for producing a rigid polyurethane foam by means of at least one blowing agent selected from the group consisting of 1,1,1,3,3-pentafluoropropane (HFC-245fa), 1,1,1,3,3-pentafluorobutane (HFC-365mfc) and a low boiling point hydrocarbon, which comprises:

at least one amine compound (1) selected from the group consisting of dimethylethylamine, dimethylpropylamine, dimethylbutylamine, dimethylpentylamine, dime

and at least one amine compound (2) selected from the group consisting of triethylenediamine, N,N,N',N'-tetramethyl-1,6-hexanediamine and N,N-dimethylcyclohexylamine,

wherein said at least one amine compound (1) and said at least one amine compound (2) are present in amounts effective to improve at least one property of a rigid polyurethane foam produced by means of said at least one blowing agent and said catalyst, compared to an otherwise identical rigid polyurethane foam produced using said at least one amine compound (1) alone, or said at least one amine compound (2) alone, as the catalyst, and wherein said at least one property includes dimensional stability.

Claim 4: The catalyst for producing a rigid polyurethane foam according to Claim 1, wherein the composition of the at least one amine compound (1) and the at least one amine compound (2) comprises from 10 to 95 wt% of the at least one amine compound (1) and from 90 to 5 wt% of the at least one amine compound (2).

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Claim 29: The catalyst for producing a rigid polyurethane foam according to Claim 1, wherein the at least one amine compound (2) comprises triethylenediamine.

Claim 30: The catalyst for producing a rigid polyurethane foam according to Claim 1, wherein the at least one amine compound (2) comprises N,N,N',N'-tetramethyl-1,6-hexanediamine.

Claim 31: The catalyst for producing a rigid polyurethane foam according to Claim 1, wherein the at least one amine compound (2) comprises N,N-dimethylcyclohexylamine.

Claim 32: The catalyst for producing a rigid polyurethane foam according to Claim 1, which additionally comprises at least one other catalyst.

Claim 33: The catalyst for producing a rigid polyurethane foam according to Claim 32, wherein the at least one other catalyst is selected from the group consisting of organic metal catalysts, metal carboxylate catalysts, tertiary amine catalysts other than amine compounds (1) and other than amine compounds (2), and quaternary ammonium salt catalysts.

Claim 37: A catalyst for producing a rigid polyurethane foam by means of at least one blowing agent selected from the group consisting of 1,1,1,3,3-pentafluoropropane (HFC-245fa), 1,1,1,3,3-pentafluorobutane (HFC-365mfc) and a low boiling point hydrocarbon, which comprises:

at least one amine compound (1) selected from the group consisting of dimethylbutylamine, dimethylbutylamine, dimethyloctylamine, and dimethyldodecylamine,

and at least one amine compound (2) selected from the group consisting of triethylenediamine, N,N,N',N'-tetramethyl-1,6-hexanediamine and N,N-dimethylcyclohexylamine,

wherein said at least one amine compound (1) and said at least one amine compound (2) are present in amounts effective to improve at least one property of a rigid polyurethane foam produced by means of said at least one blowing agent and said catalyst, compared to an otherwise identical rigid polyurethane foam produced using said at least one amine compound (1) alone, or said at least one amine compound (2) alone, as the catalyst, and wherein said at least one property includes dimensional stability.

Claim 38: A catalyst for producing a rigid polyurethane foam by means of at least one blowing agent selected from the group consisting of 1,1,1,3,3-pentafluoropropane (HFC-245fa), 1,1,1,3,3-pentafluorobutane (HFC-365mfc) and a low boiling point hydrocarbon, which comprises:

at least one amine compound (1) selected from the group consisting of dimethylethylamine, dimethylpropylamine, dimethylbutylamine, dimethylpentylamine, dime

and at least one amine compound (2) selected from the group consisting of triethylenediamine, N,N,N',N'-tetramethyl-1,6-hexanediamine and N,N-dimethylcyclohexylamine,

wherein said at least one amine compound (1) and said at least one amine compound (2) are present in amounts effective to improve a balance of properties of a rigid polyurethane foam produced by means of said at least one blowing agent and said catalyst, compared to an

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otherwise identical rigid polyurethane foam produced using said at least one amine compound (1) alone, or said at least one amine compound (2) alone, as the catalyst, and wherein said balance of properties is a combination of flowability, adhesive strength and dimensional stability.

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EVIDENCE APPENDIX

The comparative data described in the specification beginning at page 53, line 17 through page 82, line 5.

a low boiling point hydrocarbon, is used as a blowing agent, it is possible to produce a rigid polyisocyanurate foam excellent in the flame retardancy, adhesive strength and dimensional stability of the foam, without impairing the physical properties of the foam. Further, according to the process of the present invention, it is possible to obtain a rigid polyisocyanurate foam having physical properties comparable to a foam produced by a conventional process.

Now, with reference to Examples and Comparative

Examples, the catalyst for producing a rigid polyurethane
foam and the process for producing a rigid polyurethane
foam by means of such a catalyst, will be described.

However, it should be understood that the present
invention is no by means restricted by such specific
Examples.

In the following Examples and Comparative Examples, the various properties were measured by the following measuring methods.

·Measured items for the reactivity

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Cream time: The time until the foam starts to rise was measured by visual observation.

Gel time: As the reaction proceeds, the time until the liquid substance changes to a resinous substance, was measured.

Tack free time: The time until tackiness of the foam surface disappears, was measured.

Rise time: The time until the rise of the foam stops, was measured by visual observation.

•Flowability of foam: A predetermined amount of a mixed liquid was injected into an aluminum mold of $100\times25\times3.0$ cm, and the length (cm) of the formed foam was measured. The longer the length of the foam, the better the flowability.

·Core density of foam:

Free foaming was carried out by means of an aluminum mold of 50×50×4.5 cm, and the center portion of the formed foam was cut into a size of 20×20×3 cm, whereupon the size and weight were accurately measured, and core density was calculated.

·Adhesive strength of foam:

15 Forming was carried out by setting a zinc-lined iron plate of 5×5 cm on the upper side of an aluminum mold of 25×25×8.0 cm. After foaming for 1 hour, the peel strength at 90° of the set iron plate was measured and taken as the adhesive strength of the foam.

·Dimensional stability of foam:

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A foam foamed in an aluminum mold of $50\times50\times4.5$ cm was maintained under a condition of -30°C \times 48 hours, whereby a change in the thickness direction was measured. EXAMPLES 1 to 12

A polyol, a blowing agent and a foam stabilizer were mixed in a blend ratio as identified in Table 1 to prepare Premix A. 47.1 g of Premix A was taken into a

300 ml polyethylene cup, and a catalyst as identified in Table 1 was added in an amount such that the reactivity as represented by the following gel time would be 90 seconds, whereupon the temperature was adjusted to 10°C. A polyisocyanate liquid (MR-200) having the temperature adjusted to 10°C in a separate container, was put into the cup of Premix A in such an amount that the isocyanate index [isocyanate groups/OH groups (molar ratio) × 100)] would be 110, and immediately stirred by a stirrer at 6,500 rpm for 5 seconds. The mixed and stirred liquid 10 was transferred to a 2 liter polyethylene cup having the temperature adjusted to 40°C, and the reactivity during foaming was measured. Then, the starting material scale was increased, and in the same manner, the mixed liquid was put into a mold having the temperature adjusted to 40°C, whereupon foam molding was carried out. Upon expiration of 10 minutes from the introduction of the mixed liquid, the foam was removed from the mold. From the molded foam, the flowability, core density, adhesive strength and dimensional stability of the foam were 20 evaluated. The results are shown in Table 2.

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Examples 16 17 18 19 20 21 22 100 100 100 100 100 100 100 20 2.5 2.5 2.5 2.5 2.5 2.5 2.5 2.0 2.0 2.0 2.0 2.0 2.0 2.0 0.6 0.4 0.5 0.6 0.4 0.6 1.2 0.8 1.3 0.8 2X 10) 110 110 110 110 110 110		24	100		8	2.5	2.0				0.7			1.4	110
Examples 16 17 18 19 20 21 100 100 100 100 100 100 30 30 30 30 30 30 2.5 2.5 2.5 2.5 2.5 2.5 2.0 2.0 2.0 2.0 2.0 2.0 0.6 0.4 0.5 0.6 0.4 0.6 1.2		23	100		30	2.5	2.0				0.5		0.1		110
Examples 16 17 18 19 20 100 100 100 100 100 30 30 30 30 30 2.5 2.5 2.5 2.5 2.5 2.0 2.0 2.0 2.0 2.0 0.6 0.4 0.5 0.65 0.4 1.2 0.8 1.0 110 110 110		22	100		30	2.5	2.0				0.8	9.			
16 17 18 1 100 100 100 10 30 30 30 3 2.5 2.5 2.5 2 2.0 2.0 2 0.6 0.4 0.5 0.1 1.2 0.8 1.0 110 110 1	ຜ	21	100		30	2.5	2.0			9.0	•			1.2	110
16 17 18 1 100 100 100 10 30 30 30 3 2.5 2.5 2.5 2 2.0 2.0 2 0.6 0.4 0.5 0.1 1.2 0.8 1.0 110 110 1	катр1е	20	92		30	2.5	2.0			0.4	-	_	0.8		110
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16 100 30 3.5 2.5 2.0 0.6 0.6 0.6 1.2 1.2		18	100		30	2.5	2.0		0.5					1.0	110
zer ²⁾		17	100	-	8	2.5	2.0		0.4				0.8		110
Polyol " HFC-245fa HFC-365mfc Water Catalyst A 3) Catalyst B 4) Catalyst B 5) Catalyst C 6) Catalyst C 7) Catalyst C 8) Catalyst C 8)		16	100		30	2.5	2.0		9.0			1.2			110
A Amine catalyst Premix A			Polyol 1)	HFC-245fa	HFC-365mfc		Foam stabilizer ²⁾	Catalyst A 3)	Catalyst B 4)	Catalyst C 5)	Catalyst D 6)	Catalyst E "	Catalyst F 8)	Catalyst G 9)	ocyanate INDEX 10)
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nufactured by Asahi

lass Company, Limited)

Silicone type surfactant (tradename: SZ-1627), manufactured by Nippon Unicar Co.,

N, N-dimethylbutylamine (manufactured by Aldrich Co.)

N, N-dimethylhexylamine (manufactured by Aldrich Co.)

Tokyo Kasei K.K.) (manufactured by N, N-dimethyloctylamine

N, N-dimethyldodecylamine (manufactured by Tokyo Kasei K.K.)

Dipropylene glycol solution containing 33 wt% of triethylenediamine (TEDA-L33, manufactured

y TOSOH CORPORATION)

manufactured by TOSOH CORPORATION) N, N, N', N'-tetramethyl-1,6-hexanediamine (TOYOCAT-MR,

INDEX=(mols of NCO groups/mols Crude MDI (MR-200), manufactured by Nippon Polyurethane K.K., N, N-dimethylcyclohexylamine (manufactured by Aldrich Co.

f OH groups) imes 100

-1:2 143 <u>ე</u> 124 79 26 90 28.5 1.48 -1.2 122 137 14 24 8 8 1.26 29.5 -0.9 130 148 78 13 30 8 27.3 1.22 -1.5 125 138 12 82 7 9 27.5 1.28 13 123 136 83 8 2 -28.4 1.14 -0.9 139 126 10 92 8 25 27.4 1.23 -1.2 125 139 85 83 20 တ Examples 27.2 1.33 ÷ 135 122 83 8 20 ω 28.1 -0.8 1.21 128 143 79 26 9 14 27.5 1.34 극 126 8 8 21 ဖ 1.45 -1.2 27.4 122 137 20 8 8 Ŋ 28.3 1.32 -0.7 128 142 79 24 83 4 27.4 1.20 -1.2 40 125 82 8 21 က 27.3 1.29 -1.3 120 135 82 20 6 2 1.17 28.3 -1:0 129 144 25 8 8 Dimensional stability Physical properties Core density (kg/m³) Adhesive strength Reactivity (sec) Flowability (cm) Tack free time Cream time Rise time (kgf/cm^2) of foam Gel time

Table 2

28.6 1.34 -1.4 128 143 26 90 24 8 1.43 -1.2 28.7 125 138 26 8 8 -0.8 1.25 29.6 146 128 78 22 8 3 1.44 -1.2 28.7 5 126 79 25 06 2 Examples -0: 28.4 1.52 124 137 8 20 83 24 -0.7 29.3 1.30 149 130 19 78 32 91 1.57 -0 28.3 127 141 79 18 26 91 28.6 1.66 - 123 138 79 25 91 -0.7 1.42 29.5 145 128 78 16 83 29 Dimensional stability Physical properties of foam Core density (kg/m³) Adhesive strength (kgf/cm^2) Reactivity (sec) Flowability (cm) Tack free time Cream time Rise time Gel time (%)

Table 2 (continued)

COMPARATIVE EXAMPLES 1 to 7

A polyol, a blowing agent and a foam stabilizer were mixed in a blend ratio as identified in Table 3 to prepare Premix A. 47.1 g of Premix A was taken into a 300 ml polyethylene cup, and a catalyst as identified in Table 3 was added in an amount such that the reactivity as represented by the following gel time would be 90 seconds, whereupon the temperature was adjusted to 10°C. A polyisocyanate liquid (MR-200) having the temperature adjusted to 10°C in a separate container, was put into the cup of Premix A in such an amount that the isocyanate index [isocyanate groups/OH groups (molar ratio) x 100)] would be 110, and immediately stirred by a stirrer at 6,500 rpm for 5 seconds. The mixed and stirred liquid was transferred to a 2 liter polyethylene cup having the 15 temperature adjusted to 40°C, and the reactivity during foaming was measured. Then, the starting material scale was increased, and in the same manner, the mixed liquid was put into a mold having the temperature adjusted to 40°C, whereupon foam molding was carried out. Upon 20 expiration of 10 minutes from the introduction of the mixed liquid, the foam was removed from the mold. From the molded foam, the flowability, core density, adhesive strength and dimensional stability of the foam were evaluated. The results are shown in Table 4. 25

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-	14	100		30	2.5	2.0							1.8	=	δ
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	12	001		30	2.5	2.0					1.9			110	ufact
	11	901	,	9	2.5	2.0				3.1				110	
	10	100		90	2.5	2.0			2.6					110	mgKOH/g,
mples	6	100		90	2.5	2.0		2.3				•		110	0 mgK
Comparative Examples	ω	100		90	2.5	2.0	2.0			-				110	value=440
ativ	7	100	99		2.5	2.0							2.8	110	l val
ompar	9	100	30		2.5	2.0			٠.		•	2.1		110	1 (OF
Ū	.C	901	30		2.5	2.0					2.9			110	polyether polyol (OH
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		Polyol 1)	× HFC-245fa	HFC-365mfc	Water	Foam stabilizer	Cata]	sY.			g Catalyst E "	تا Catalyst F	R Catalyst G 9)	Isocyanate INDEX 10)	Sucrose/aromatic ami
			Blend (parts by weight)][

Company, Limited) sahi

Ltd SZ-1627), manufactured by Nippon Unicar Co. by Aldrich Co.) (tradename: type surfactant Silicone

Aldrich Co. (manufactured by manufactured N, N-dimethylbutylamine N, N-dimethylhexylamine

Tokyo Kasei K.K. (manufactured by N, N-dimethyloctylamine

N,N-dimethyldodecylamine (manufactured by Tokyo Kasei K.K.)

Dipropylene glycol solution containing 33 wt% of triethylenediamine (TEDA-L33,

nanufactured by TOSOH CORPORATION)

N,N,N',N'-tetramethy1-1,6-hexanediamine (TOYOCAT-MR, manufactured by TOSOH CORPORATION) N, N-dimethylcyclohexylamine (manufactured by Aldrich Co.)

(MR-200), manufactured by Nippon Polyurethane K.K., INDEX=(mols of NCO groups/mols of OH groups) × 100 Crude MDI

28.6 146 133 25 9/ -4.4 9 13 137 148 28.5 0.76 -4.3 24 8 75 136 150 29.7 0.67 -2.7 8 9 72 28.6 144 160 1.55 -7.2 26 8 9/ <u>0</u> 150 28.4 -7.8 162 1.68 25 8 75 Comparative Examples 28.5 145 1.75 -7.5 154 23 89 75 O , 168 28.6 155 1.62 -8.2 8 24 75 ∞ 27.5 0.48 -5.6 140 127 20 91 9/ 27.4 130 142 0.51 -5.2 20 8 9/ 9 28.6 0.45 129 143 -3.5 25 8 72 Ŋ 27.3 137 153 1.41 -7.7 8 9/ 21 4 27.5 -8.5 143 1.50 154 90 75 21 က 138 27.4 147 1.56 -9.2 20 9 0 77 27.5 1.38 -8.8 160 141 20 8 9/ Dimensional stability Physical properties Core density (kg/m³) Adhesive strength Reactivity (sec) Flowability (cm) Tack free time Cream time Rise time Gel time of foam (kgf/cm^2) %

EXAMPLES 13 to 24

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A polyol, a blowing agent and a foam stabilizer were mixed in a blend ratio as identified in Table 1 to prepare Premix A. 47.1 g of Premix A was taken into a 300 ml polyethylene cup, and a catalyst as identified in Table 1 was added in an amount such that the reactivity as represented by the following gel time would be 90 seconds, whereupon the temperature was adjusted to 10°C. A polyisocyanate liquid (MR-200) having the temperature adjusted to 10°C in a separate container, was put into the cup of Premix A in such an amount that the isocyanate index [isocyanate groups/OH groups (molar ratio) \times 100)] would be 110, and immediately stirred by a stirrer at 6,500 rpm for 5 seconds. The mixed and stirred liquid was transferred to a 2 liter polyethylene cup having the temperature adjusted to 40°C, and the reactivity during foaming was measured. Then, the starting material scale was increased, and in the same manner, the mixed liquid was put into a mold having the temperature adjusted to 40°C, whereupon foam molding was carried out. Upon expiration of 10 minutes from the introduction of the mixed liquid, the foam was removed from the mold. From the molded foam, the flowability, core density, adhesive strength and dimensional stability of the foam were evaluated. The results are shown in Table 2. COMPARATIVE EXAMPLES 8 to 14

A polyol, a blowing agent and a foam stabilizer were

mixed in a blend ratio as identified in Table 3 to prepare Premix A. 47.1 g of Premix A was taken into a 300 ml polyethylene cup, and a catalyst as identified in Table 3 was added in an amount such that the reactivity as represented by the following gel time would be 90 seconds, whereupon the temperature was adjusted to 20°C. A polyisocyanate liquid (MR-200) having the temperature adjusted to 20°C in a separate container, was put into the cup of Premix A in such an amount that the isocyanate index [isocyanate groups/OH groups (molar ratio) × 100)] would be 110, and immediately stirred by a stirrer at 6,500 rpm for 5 seconds. The mixed and stirred liquid was transferred to a 2 liter polyethylene cup having the temperature adjusted to 40°C, and the reactivity during foaming was measured. Then, the starting material scale was increased, and in the same manner, the mixed liquid was put into a mold having the temperature adjusted to 40°C, whereupon foam molding was carried out. Upon expiration of 10 minutes from the introduction of the mixed liquid, the foam was removed from the mold. From the molded foam, the flowability, core density, adhesive strength and dimensional stability of the foam were evaluated. The results are shown in Table 4. COMPARATIVE EXAMPLES 15 to 29

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A polyol, a blowing agent and a foam stabilizer were mixed in a blend ratio as identified in Table 5 to prepare Premix A. 47.1 g of Premix A was taken into a

300 ml polyethylene cup, and a catalyst as identified in Table 5 was added in an amount such that the reactivity as represented by the following gel time would be 90 seconds, whereupon the temperature was adjusted to 20°C. A polyisocyanate liquid (MR-200) having the temperature adjusted to 20°C in a separate container, was put into the cup of Premix A in such an amount that the isocyanate index [isocyanate groups/OH groups (molar ratio) × 100)] would be 110, and immediately stirred by a stirrer at 6,500 rpm for 5 seconds. The mixed and stirred liquid 10 was transferred to a 2 liter polyethylene cup having the temperature adjusted to 40°C, and the reactivity during foaming was measured. Then, the starting material scale was increased, and in the same manner, the mixed liquid was put into a mold having the temperature adjusted to 15 40°C, whereupon foam molding was carried out. Upon expiration of 10 minutes from the introduction of the mixed liquid, the foam was removed from the mold. the molded foam, the flowability, core density, adhesive strength and dimensional stability of the foam were evaluated. The results are shown in Table 6.

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		15	16	17	18	19	20	21	22	23	24	25	26	27	28	23
Polvol 1	1)	92	100	5	8	50	90	90	100	100	901	100	001	8	90	8
HCFC	HCFC-141b	25	25	25	25	25	52	25	25	25.	25	25	25	25	22	22
A Water	· h	2.5	2.5	2.5	2.5	2.5	2.0	5.0	2.0	2.0	2.0	2.0	2.0	2.0	5.0	2.0
Foam	Foam stabilizer 2)	2.0	2.0	2.0	2.0	2.0	2.0	2.0	2.0	2.0	2.0	2.0	2.0	2.0	2.0	2.0
Cata	Catalyst A 3)	0.4	0.3	0.4			• •									
Cata	Catalyst B 4)				0.5	0.35	0.45									
Cate	Catalvst C ⁵⁾							0.55	0.35	0.5						•
18: Ca Ta	Catalyst D 6)				·						0.65	0.45	9.0			
Cate	Catalyst E "	0.8			1.0			Ξ			1.3			1.5		
Cate	Catalyst F 8)		9.0			0.7			0.7			0.9			1.2	
Cate	Catalyst G 9)	_	_	0.8			0.9			1.0			1.2			1.6
\ \rac{1}{2}	Tsocvanate INDEX 10)	2	110	110	19	110	110	110	110	110	9	110	110	110	9=	110
3						,	1	2 / LIO/1	7 / 0	1/4/	~/ FIC		1004.	raniforting hy Acabi	7	ירלבם.

Sucrose/aromatic amine type polyether polyol (OH value=440 mgKOH/g, manufactured

Glass Company, Limited)

Silicone type surfactant (tradename: SZ-1627), manufactured by Nippon Unicar Co., (manufactured by Aldrich Co.

Aldrich Co. N, N-dimethylbutylamine

Tokyo Kasei K.K.) (manufactured by (manufactured by N, N-dimethyloctylamine N, N-dimethylhexylamine

N,N-dimethyldodecylamine (manufactured by Tokyo Kasei K.K.) Dipropylene glycol solution containing 33 wt% of triethylenediamine (TEDA-L33, manufactured by TOSOH CORPORATION)

8) N,N',N'-tetramethyl-1,6-hexanediamine (TOYOCAT-MR, manufactured by TOSOH CORPORATION) N, N-dimethylcyclohexylamine (manufactured by Aldrich Co.)

INDEX=(mols of NCO groups/mols Crude MDI (MR-200), manufactured by Nippon Polyurethane K.K.,

						Comp	Comparative		Examples	les					
	15	16	17	8-	19	20	21	22	23	24	25	26	27	28	29
Reactivity (sec)															
Cream time	31	25	27	29	24	22	<u>ب</u>	25	25	30	25	56	<u>ਦ</u>	25	56
Gel time	<u>6</u>	68	06	8	9.	6	06	06	6	91	06	06	06	.06	06
Tack free time	131	123	125	129	122	126	130	124	125	128	126	125	129	125	126
Rise time	146	139	142	143	137	140	145	138	139	143	136	139	143	139	140
Physical properties of foam								-							
Flowability (cm)	8	84	83	8	83	82	~~~	83	83	8	84	83	81	84	84
Core density (kg/m³)	29.5	28.2	28.4	29.3	28.3	28.0	29.0	28.2	28.4	29.3	28.4	28.3	29.3	28.0	28.1
Adhesive strength (kgf/cm²)	1.62	1.76	1.71	1.70	1.86	1.79	1.67	1.82	1.75	1.65	1.83	1.72	1.62	1.78	1.70
Dimensional stability (%)	-1.0	-1.2	-1.3	-0.8	-1.2	-1.1	-0.8	-1.0	-1.2	-0.9	-1.3	-1.5	-0.9	-1.3	-1.2

As is evident from Tables 2, 4 and 6, it is possible to produce foams excellent in flowability, adhesive strength and dimensional stability by using amine compounds of the present invention as catalysts.

Namely, Examples 1 to 12 are examples wherein rigid polyurethane foams were produced by using catalysts of the present invention and 1,1,1,3,3-pentafluoropropane (HFC-245fa) as a blowing agent. In each Example, a rigid urethane foam excellent in flowability, adhesive strength and dimensional stability, was obtained. Further, Examples 13 to 24 are Examples wherein rigid polyurethane foams were produced by using the catalysts of the present invention and 1,1,1,3,3-pentafluorobutane (HFC-365mfc) as a blowing agent. In each Example, a rigid urethane foam excellent in flowability, adhesive strength and dimensional stability, was obtained.

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Whereas, Comparative Examples 1 to 4 and Comparative Examples 8 to 11 are examples wherein rigid polyurethane foams were produced by using only the aliphatic amine compounds of the above formula (1) as catalysts, whereby large amounts of the catalysts were required, and the foams were inferior in flowability and dimensional stability.

Further, Comparative Examples 5 to 7 and Comparative

Examples 12 to 14 are examples wherein rigid polyurethane
foams were produced by using triethylenediamine,

N,N,N',N'-tetramethyl-1,6-hexanediamine or N,N-

dimethylcyclohexylamine solely as the catalyst without using the aliphatic amine compound of the above formula (1), whereby the obtained foams were inferior in flowability, adhesive strength and dimensional stability.

Further, Comparative Examples 15 to 21 are examples in which 1,1-dichloro-1-fluoroethane (HCFC-141b) was used as a blowing agent, whereby even if the catalyst of the present invention was used, no remarkable effect was observed in the flowability, adhesive strength and dimensional stability of the foams.

EXAMPLES 25 to 48

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A polyol, a blowing agent and a foam stabilizer were mixed in a blend ratio as identified in Table 7 to prepare Premix A. 47.1 g of Premix A was taken into a 300 ml polyethylene cup, and a catalyst as identified in Table 7 was added in an amount such that the reactivity as represented by the following gel time would be 90 seconds, whereupon the temperature was adjusted to 20°C. A polyisocyanate liquid (MR-200) having the temperature adjusted to 10°C in a separate container, was put into the cup of Premix A in such an amount that the isocyanate index [isocyanate groups/OH groups (molar ratio) \times 100)] would be 110, and immediately stirred by a stirrer at 6,500 rpm for 5 seconds. The mixed and stirred liquid was transferred to a 2 liter polyethylene cup having the temperature adjusted to 40°C, and the reactivity during foaming was measured. Then, the starting material scale

was increased, and in the same manner, the mixed liquid was put into a mold having the temperature adjusted to 40°C, whereupon foam molding was carried out. Upon expiration of 10 minutes from the introduction of the mixed liquid, the foam was removed from the mold. From the molded foam, the flowability, core density, adhesive strength and dimensional stability of the foam were evaluated. The results are shown in Table 8.

_									<u> </u>							\neg
	- }	37	80	50	0	4	1.5	1.5	0.35			<u> </u>	0.70			110
	<u> </u>	36	. 08	20	91		.5	1.5				0.55			1.10	110
		35	8	20	16		1.5	1.5				0.40		0.80		92
		34	80	20	91		.5	1.5				09.0	1.20			110
	- '	33	80	50	91		1.5	1.5			0.45	,			06.0	110
	Ø	32	88	20	91		1.5	1.5			0.33			0.65		110
	Examples	31	8	70	91		1.5	1.5			0.50		00.1			110
	EX	8	8	70	16		5.1	.5	٠.	0.80					1.60	110
		29	8	70	16		1.5	1.5		0.30			•	09.0		110
		28	8	20	16		5.	1.5		0.45			0.90			110
		27	8	70	- 91		.5	1.5	0.35						0.70	110
		26	8	20	16		1.5	1.5	0.25					0.50		110
		25	8	50	16		1.5	1.5	0.38				0.75			110
7			1) ه امبرامو	Polyol B 2)	Cvclopentane	Teopontano		Foam stabilizer 3)	Catalyst A 4)				Catalvst	Catalvst	1 a d	
Table 7					A x	ţшe						.ats		utw	Ą	SI
ľak	1					(=	цф	i əw	ρλ	sj.	bэт) p	цəт	B		

(continued) Table

				•			EX	Examples	S					
			38	39	40	41	42	43	44	45	46	47	48	
		Polvol A 1)	8	8	8	8	8	8	80	. 80	8	8	08	
	A		70	20	20	50	20	50	70	70	20	50	70	
(41	хŢ	Cyclopentane	10	9	9	0	2	9	9	9	9	9	2	
дБ ⁻	ш ә	Isopentane	4	4	4	4	4	4	4	4	4	4	4	
ţə/	Σđ	Water		1.5	5.	1.5	1.5	7.	1.5	. 5.	5.	. .	5.	
w Y		Foam stabilizer 3)	1.5	1.5	1.5	1.5	1.5	1.5	1.5	1.5	1.5	1.5	1.5	
ſq] 7	Catalyst A 4)	0.23	0.33										
87	SV.	Catalyst B 5)		_	0.40	0.28	0.70							
gr		Gatalyst C 6)						0.45	0.30	0.40				
đ)		Catalvst D 7)									0.55	0.35	0.50	
рu) ə	Catalyst E 8)			08.0			06.0			1.10			
ŢĢī	uŢ	Gatalyst F 9)	0.45			0.55			09.0			0.70		
В	шĄ	Catalyst G 10)		0.65		-	1.40			0.80			9.	
		Isocvanate INDEX 11)	2	110	110	110	110	110	110	110	110	10	=	
1	<u> </u> 3	1) Waste PET type polyester		701 ((polyol (OH value=241	lue=2		mgKOH/g)		manufactured		by Ox	by Oxid Co	

Mannich type polyether polyol (OH value=469 mgKOH/g) manufactured by Daiichi Kogyo

L-5420), manufactured by Nippon Unicar Co. (manufactured by Aldrich Co.) Silicone type surfactant (tradename:

N, N-dimethylbutylamine

(manufactured by Tokyo Kasei K.K.) N, N-dimethylhexylamine (manufactured by Aldrich Co.

N, N-dimethyloctylamine

N, N-dimethyldodecylamine (manufactured by Tokyo Kasei K.K.)

solution containing 33 wt% of triethylenediamine (TEDA-L33, manufactured by TOSOH CORPORATION) Dipropylene glycol

9) N,N,N',N'-tetramethyl-1,6-hexanediamine (TOYOCAT-MR, manufactured by TOSOH CORPORATION) N, N-dimethylcyclohexylamine (manufactured by Aldrich Co.)

INDEX=(mols of NCO Crude MDI (MR-200), manufactured by Nippon Polyurethane K.K., groups/mols of OH groups) imes 100

25 26 27 28 29 30 31 32 33 34 35 30 25 26 29 25 26 31 25 25 30 26 90 90 91 90 91 90 91 90 150 143 143 143 143 144 148 145 161 147 153 155 148 151 158 148 150 150 78 79 80 76 78 76 80 76 80 164 181 168 1.85 2.03 1.88 1.69 1.72 1.6 1.79	Table 8												
25 26 27 28 29 30 31 25 25 25 30 26 31 55 48 148 150 143 144 148 148 145 148 151 158 151 158 151 158 151 158 151 158 151 158 151 158 151 158 151 158 151 158 151 151			-	!			Exam	oles					
10 25 26 29 25 26 31 25 25 30 26 30 30 30 30 30 30 30 30 30 30 30 30 30		25	26	27	28	59	30	31	32	33	34	35	36
130 25 26 29 25 26 31 25 25 30 26 30 30 30 30 30 30 30 30 30 30 30 30 30	Reactivity (sec)										,		-
f foam [150] 143 143 148 143 150 148 148 149 149 149 145 145 145 150 143 141 148 145 148 145 148 145 148 145 148 145 148 145 148 145 148 145 148 148 148 148 148 148 148 148 148 148	Cream time	30	25	792	53	25	56	31	25	25	30	56	56
f f oam 150 143 148 143 163 155 148 151 158 148 150 144 148 145 148 f f oam 161 147 153 155 148 151 158 148 150 157 148 f f coam 78 78 78 76 80 76 80 76 80 31.6 30.4 30.6 31.6 30.7 31.3 30.3 30.6 31.7 30.7 cyf/cm² 1.64 1.81 1.68 1.85 2.03 1.88 1.69 1.72 1.6 1.79 cy (96) -0.9 -1.2 -1.1 -0.6 -1.1 -1.0 -0.7 -1.0 -1.1 -0.9 -1.2 -1.1 -0.9 -1.1 -0.7 -1.0 -1.1 -0.7 -1.0 -1.1 -0.7 -1.0 -1.1 -1.2 -1.1 -1.1 -0.7 -1.0 -1.1 <td< td=""><td>Gel time</td><td>06</td><td>06</td><td>91</td><td>90</td><td>68</td><td>06</td><td>91</td><td>06</td><td>06</td><td>91</td><td>06</td><td>90</td></td<>	Gel time	06	06	91	90	68	06	91	06	06	91	06	90
f foam 161 147 153 155 148 151 158 148 150 157 148 f foam 78 78 76 80 79 76 80 31.6 30.4 30.6 31.6 30.7 31.3 30.3 30.6 31.7 30.7 cyt(cm²) 1.64 1.81 1.68 1.85 2.03 1.88 1.69 1.72 1.6 1.79 cyt(cw) -0.9 -1.2 -1.1 -0.6 -1.1 -1.0 -0.7 -1.0 -1.1 -0.8 -1.2 -1.1 -0.0 -1.1 -0.7 -1.0 -0.7 -1.0 -0.1 -0.1 -0.8 -1.2	Tack free time	150	143	143	148	143	143	150	143	144	148	145	144
f foam 78 79 80 76 78 78 76 80 79 76 80 30.7 31.3 30.3 30.6 31.7 30.7 30.7 1.64 1.81 1.68 1.85 2.03 1.88 1.69 1.86 1.72 1.6 1.79 1.2	Rise time	161	147	153	155	148	151	158	148	150	157	148	153
78 79 80 76 78 76 80 79 76 80 31.6 30.4 30.6 31.6 30.7 31.3 30.3 30.6 31.7 30.7 1gf/cm² 1.64 1.81 1.68 1.85 2.03 1.88 1.69 1.86 1.72 1.6 1.79 Ly 96) -0.9 -1.2 -1.1 -0.6 -1.1 -1.0 -0.7 -1.0 -1.1 -0.8 -1.2	44												
h (kgf/cm²) 31.6 30.4 30.6 31.6 30.6 30.7 31.3 30.3 30.6 31.7 30.7 30.7 h (kgf/cm²) 1.64 1.81 1.68 1.85 2.03 1.88 1.69 1.86 1.72 1.6 1.79 1.15	Flowability (cm)	78	79	. 80	9/	28	78	9/	80	79	9/	80	79
gf/cm²) 1.64 1.81 1.68 1.85 2.03 1.88 1.69 1.86 1.72 1.6 1.79 1.79 1.79 1.79 1.79 1.79 1.79 1.79	Core density (kg/m³)	31.6	30.4	30.6	31.6	30.6	30.7	31.3	30.3	30.6	31.7	30.7	30.4
-0.9 -1.2 -1.1 -0.6 -1.1 -1.0 -0.7 -1.0 -1.1 -0.8 -1.2	Adhesive strength (kgf/cm ²)	1.64	18.1	1.68	1.85	2.03	1.88	1.69	1.86	1.72	1.6	1.79	1.7
	Dimensional stability (%)	-0.9	-1.2	7	-0.6	-1.1	-1.0	-0.7	-1.0	1.1	-0.8	-1.2	-1.4

29.5 1.66 144 148 8 25 8 8 1.74 29.8 = 143 146 90 25 8 47 1.55 30.8 -0.7 149 158 11 29 46 9 -1.0 29.7 1.67 150 143 8 24 83 45 1.80 29.4 -0.9 143 145 8 24 90 44 1.64 30.4 9.0-149 155 Examples 11 43 8 8 -0.9 1.82 29.8 143 151 79 25 8 42 1.97 -1.0 . 06 29.7 142 148 79 24 4 -0.5 30.7 1.80 148 159 8 28 9 -1.0 29.7 1.63 145 150 . 52 8 8 39 29.5 1.76 - 145 140 24 8 8 38 **-0.8** 30.7 Adhesive strength (kgf/cm^2) 1.59 162 151 79 29 83 37 Dimensional stability (%) Physical properties of foam Core density (kg/m^3) Flowability (cm) Tack free time Reactivity (sec) Cream time Rise time Gel time

Table 8 (continued)

COMPARATIVE EXAMPLES 30 to 43

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A polyol, a blowing agent and a foam stabilizer were mixed in a blend ratio as identified in Table 9 to prepare Premix A. 47.1 g of Premix A was taken into a 300 ml polyethylene cup, and a catalyst as identified in Table 9 was added in an amount such that the reactivity as represented by the following gel time would be 90 seconds, whereupon the temperature was adjusted to 20°C. A polyisocyanate liquid (MR-200) having the temperature adjusted to 20°C in a separate container, was put into the cup of Premix A in such an amount that the isocyanate index [isocyanate groups/OH groups (molar ratio) × 100)] would be 110, and immediately stirred by a stirrer at 6,500 rpm for 5 seconds. The mixed and stirred liquid was transferred to a 2 liter polyethylene cup having the temperature adjusted to 40°C, and the reactivity during foaming was measured. Then, the starting material scale was increased, and in the same manner, the mixed liquid was put into a mold having the temperature adjusted to 40°C, whereupon foam molding was carried out. Upon expiration of 10 minutes from the introduction of the mixed liquid, the foam was removed from the mold. From the molded foam, the flowability, core density, adhesive strength and dimensional stability of the foam were evaluated. The results are shown in Table 10. 25

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							compa.	comparative	e Exa	Examples	70				
· [30	31	32	33	34	35	36	37	38	39	40	41	42	43
	Polyol A 1)	8	80	80	80	80	08	02	ç	8	Q	ď	6	9	2 8
A	A Polvol B 2)	20	- 6	ç	ç	8	3 8	3 8	3	3 :	3	8	0	2	<u></u>
x		· 2	3	3	7	7	07	2.5	70	50	70	70	20	20	20
Ţu	- Cyclopentane	16	91	16	16	16	91	16	2	10	9	10	0	E	10
æ.	Isopentane								4	4	4	4	. 4	: <	: <
ιď	Water	1.5	1.5	1.5	.5	1.5	5:	.5	7.	. <u>.</u>	. <u>.</u>	. t	٠ ـ	- L	+ 4
	Foam stabilizer	3) 1.5	-5.	1.5	1.5	1.5	1.5	.5	.5	. rc	<u>.</u>	. rc	<u>.</u>		
75	Catalyst A 4)	1.60							145				:	2	3
Ţλε	Catalyst B ⁵⁾		1.85					•		165				,	
£З	Catalyst C 6)	-		2.10						2	0	-			
cs	Catalyst D "				2.55						3	2 30			
əτ	Catalyst E 8)					1.55		<u> </u>				3	1 40		
ıţw	Catalyst F 9)						1.10				_		?	1 05	
A	Catalyst G ¹⁰⁾							1.45	_	-				3	1 30
Š	Isocyanate INDEX 11)	110	110	110	110	110	110	110	110	4	110	110	5	5	3 5
3	Macto DET time noline	1001				1	7	2	- - -	2	2	2	2	2	110

Mannich type polyether polyol (OH value=469 mgKOH/g) manufactured by Daiichi Kogyo type polyester polyol (OH value=241 mgKOH/g) manufactured by Oxid Co.

Seiyaku Co., Ltd.

Silicone type surfactant (tradename: L-5420), manufactured by Nippon Unicar Co.,

N, N-dimethylbutylamine

(manufactured by Aldrich Co.) N, N-dimethylhexylamine

Tokyo Kasei K.K. (manufactured by N, N-dimethyloctylamine

solution containing 33 wt% of triethylenediamine (TEDA-L33 N, N-dimethyldodecylamine (manufactured by Tokyo Kasei K.K. Dipropylene glycol

manufactured by TOSOH CORPORATION)

9) N,N,N',N'-tetramethyl-1,6-hexanediamine (TOYOCAT-MR, manufactured by TOSOH CORPORATION) N, N-dimethylcyclohexylamine (manufactured by Aldrich Co.)

Crude MDI (MR-200), manufactured by Nippon Polyurethane K.K., INDEX=(mols of NCO groups/mols of OH groups) imes 100

					ပြ	mpar	Comparative		Examples	S C				
	30	31	32	33	34	35	36	37	38	39	9	14	42	43
Reactivity (sec)						,								
Cream time	25	24	26	25	30	25	56	24	24	25	24	29	24	25
Gel time	06	91	06	06	68	06	06	06	06	. 06	91	68	06	06
Tack free time	145	143	148	142	150	140	143	143	143	146	140	151	143	141
Rise time	156	157	159	153	162	. 155	160	155	156	157	155	160	158	156
Physical properties of foam													-	
Flowability (cm)	73	74	72	73	02	73	73	74	75	73	74	0/	74	74
Core density (kg/m³)	32.2	32.1	32.2	32.0	33.5	32.1	32.2	31.9	31.8	31.9	31.6	33.2	31.8	31.9
Adhesive strength (kgf/cm²)	1.79	2.03	1.95	1.83	0.90	1.02	96.0	1.74	1.95	1.88	1.78	0.88	1.00	0.95
Dimensional stability (%)	-7.7	-8.1	-7.5	8.9-	-3.1	-4.6	-5.0	-6.8	-7.2	-6.8	-6.0	-2.8	-4.3	-4.3

COMPARATIVE EXAMPLES 44 to 58

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A polyol, a blowing agent and a foam stabilizer were mixed in a blend ratio as identified in Table 11 to prepare Premix A. 47.1 g of Premix A was taken into a 300 ml polyethylene cup, and a catalyst as identified in Table 11 was added in an amount such that the reactivity as represented by the following gel time would be 90 seconds, whereupon the temperature was adjusted to 20°C. A polyisocyanate liquid (MR-200) having the temperature adjusted to 20°C in a separate container, was put into the cup of Premix A in such an amount that the isocyanate index [isocyanate groups/OH groups (molar ratio) × 100)] would be 110, and immediately stirred by a stirrer at 6,500 rpm for 5 seconds. The mixed and stirred liquid was transferred to a 2 liter polyethylene cup having the temperature adjusted to 40°C, and the reactivity during foaming was measured. Then, the starting material scale was increased, and in the same manner, the mixed liquid was put into a mold having the temperature adjusted to 40°C, whereupon foam molding was carried out. Upon expiration of 10 minutes from the introduction of the mixed liquid, the foam was removed from the mold. From the molded foam, the flowability, core density, adhesive strength and dimensional stability of the foam were evaluated. The results are shown in Table 12.

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	58	8	20	22	1.5	1.5							1.45	110	
	57	8	20	25	1.5	1.5						1.10		110	
	56	8	20	25	1.5	1.5					1.35			110	
	55	80	70	25	5.	1.5				0.55			1.10	110	
	54	80	20	25	1.5	1.5				0.40		0.80		110	
ole 1	53	8	20	25	5.	1.5				09.0	1.20			110	
Comparative Example	52	8	20	25	1.5	1.5			0.45				0.00	92	
ive]	51	8	70	25	5.	1.5		•	0.30			0.60		110	_
barat	20	8	20	25	5.	1.5			0.50		1.0			110	
Comr	49	8	70	25	5.	1.5		0.40					0.80	110	
	48	8	20	25	.5	1.5		0.33				0.65		110	
	47	8	70	25	5.	1.5		0.45			0.90			110	
	46	8	70	25	5.	1.5	0.35						0.70	110)
	45	8	20	25	1.5	.5	0.25					0.50		E C	- -
ļ ŧ	44	8	2	25	. 5	5.	0.35				0.70			110	<u> </u>
		1) کا امریاط	Dolvol B 2)	THUEST THE	Mater	Foam stabilizer 3)	Catalyst A 4)	Catalvst B 5)	Gatalvst C 6)	Catalyst D 7)	Catalyst E 8)	Catalvst F 9)	A Catalvet G 10)	Tacarranto TNDEV 11)	くいつれていていてい
		-	A :	χŢΊ	GT.	ew By	F pX	5/	בב	ьч 16:) F	u i	BJ.		=
			- (-44	'nί	Q (A)	<i>1</i> 14	_ =	<u> </u>		<u>, </u>		La		_

Mannich type polyether polyol (OH value=469 mgKOH/g) manufactured by Daiichi Kogyo Waste PET type polyester polyol (OH value=241 mgKOH/g) manufactured by Oxide

Seiyaku Co., Ltd.

L-5420), manufactured by Nippon Unicar Co., Ltd manufactured by Aldrich Co. Silicone type surfactant (tradename: N, N-dimethylbutylamine

(manufactured by Aldrich Co. N, N-dimethylhexylamine

Tokyo Kasei K.K. (manufactured by N, N-dimethyloctylamine

N, N-dimethyldodecylamine (manufactured by Tokyo Kasei K.K.

Dipropylene glycol solution containing 33 wt% of triethylenediamine (TEDA-L33,

9) N,N,N',N'-tetramethyl-1,6-hexanediamine (TOYOCAT-MR, manufactured by TOSOH CORPORATION manufactured by TOSOH CORPORATION)

INDEX= (mols of NCO Crude MDI (MR-200), manufactured by Nippon Polyurethane K.K. N, N-dimethylcyclohexylamine (manufactured by Aldrich Co.)

groups/mols of OH groups) $\times~100$

29.3 1.72 -1.3 1.80 -1.4 29.2 <u>8</u> 1.64 -1.0 30.5 29.5 -1.5 1.85 1.74 -1.4 29.6 1.77 | 1.67 -0.9 30.5 Comparative Examples 29.6 ۔ ن 29.4 -1.69 | 1.84 -0.9 30.2 29.2 1.81 -0 1.88 -1.3 29.5 1.72 30.5 -0.9 1.73 29.6 -1.5 <u>5</u> 1.78 29.4 -1.4 30.4 1.64 Physical properties Core density (kg/m³) Adhesive strength Reactivity (sec) Flowability (cm) Tack free time stability (%) Dimensional Cream time Rise time of foam (kgf/cm^2) Gel time

As is evident from Tables 8, 10 and 12, it is possible to produce foams excellent in flowability, adhesive strength and dimensional stability by using amine compounds of the present invention as catalysts.

Namely, Examples 25 to 48 are examples wherein rigid polyurethane foams were produced by using low boiling point hydrocarbons having boiling points of from -30 to 90°C as blowing agents. In each Example, a rigid urethane foam excellent in flowability, adhesive strength and dimensional stability, was obtained.

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Whereas, Comparative Examples 30 to 33 and Comparative Examples 37 to 40 are examples wherein rigid polyurethane foams were produced by using only the aliphatic amine compounds of the above formula (1) as catalysts, whereby large amounts of the catalysts were required, and the foams were inferior in flowability and dimensional stability.

Further, Comparative Examples 34 to 36 and Comparative Examples 41 to 43 are examples wherein rigid polyurethane foams were produced by using triethylenediamine, N,N,N',N'-tetramethyl-1,6-hexanediamine or N,N-dimethylcyclohexylamine solely as the catalyst without using the aliphatic amine compound of the above formula (1), whereby the foams were inferior in flowability, adhesive strength and dimensional stability.

Further, Comparative Examples 44 to 58 are examples

in which 1,1-dichloro-1-fluoroethane (HCFC-141b) was used as a blowing agent, whereby even if the catalyst of the present invention was used, no distinct effect to the flowability, adhesive strength and dimensional stability, was observed.

EXAMPLES 49 to 52 and COMPARATIVE EXAMPLES 59 to 61

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A polyol, a blowing agent and a foam stabilizer were mixed in a blend ratio as identified in Table 13 to prepare Premix A. 47.1 g of Premix A was taken into a 300 ml polyethylene cup, and a catalyst as identified in Table 13 was added in an amount such that the reactivity as represented by the following gel time would be 90 seconds, whereupon the temperature was adjusted to 10°C. A polyisocyanate liquid (MR-200) having the temperature adjusted to 10°C in a separate container, was put into the cup of Premix A in such an amount that the isocyanate index [isocyanate groups/OH groups (molar ratio) \times 100)] would be 110, and immediately stirred by a stirrer at 6,500 rpm for 5 seconds. The mixed and stirred liquid was transferred to a 2 liter polyethylene cup having the temperature adjusted to 40°C, and the reactivity during foaming was measured. Then, the starting material scale was increased, and in the same manner, the mixed liquid was put into a mold having the temperature adjusted to 40°C, whereupon foam molding was carried out. Upon expiration of 10 minutes from the introduction of the mixed liquid, the foam was removed from the mold. From

Application No. 10/618,693 Corrected Appeal Brief

RELATED PROCEEDINGS APPENDIX

None.